Many of the most relevant chemical properties of matter depend explicitly on atomistic details, rendering an atomistic resolution mandatory. Alas, even when using high-performance computing, brute force high-throughput screening of compounds is beyond any capacity for all but the simplest systems and properties due to the combinatorial nature of chemical space, i.e. all the compositional, constitutional, and conformational isomers. Consequently, an efficient computational compound design algorithm must not only make a trade-off between sufficient accuracy of applied models and computational speed, but must also aim for rapid convergence in terms of number of compounds visited. In this talk, I will first discuss the coupling of molecules through the use of fractional nuclear charges, and how one can calculate the corresponding "alchemical" property gradients in chemical compound space. Thereafter, I will describe recently developed statistical approaches for interpolating (Kriging) quantum mechanical observables in chemical space. For both methods examples will be presented for predicting properties of out-of-sample molecules with "chemical accuracy" at a fraction of the computational cost.